On the magnetic instability of ultrathin fcc Fe_xNi_{1-x} films

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The 'invar effect' in Fe_xNi_{1-x} alloys occurs when the Fe content approaches 65%. At this point, the magnetization falls to zero, and a martensitic structural transformation from a fcc to a bcc lattice occurs. This paper addresses the question: "What happens if this structural transformation is suppressed in an ultrathin alloy film?"

In the bulk, Fe_xNi_{1-x} alloys show anomalous behavior at a Fe content of ~65%, which is usually referred to as the 'invar effect'[1]. It has been observed that the structure changes from the fcc into the bcc phase as the Fe content increases. Simultaneously the Curie temperature T_c and the magnetic moment collapse[1]. The question arises to what extent the magnetic properties are modified if the fcc phase stability is extended beyond 65% Fe. From a theoretical point of view it is predicted that a moment collapse still takes place at a Fe content of 75%, which is accompanied by a reduction of the atomic volume of ~9% [2]. Experimentally an extended regime of fcc stability can be achieved via epitaxy on a Cu(100) substrate [3].We report here on the correlation of the magnetic dichroism of the Fe 3p core level in photoemission with linearly polarized light (LMDAD) to the atomic volume[4]. In particular we focus on the thickness dependence for Fe concentrations around 75%.

Details of sample preparation can be found elsewhere[3]. The dichroism experiments were performed at the SpectroMicroscopy Facility on Beamline 7 at the Advanced Light Source, Berkeley [5].For photoemission of the 3p core levels we utilized 190 eV photons (p-polarized) and collected electrons in normal emission more details have been published elsewhere[6].We have demonstrated that LMDAD measures a quantity closely related to the element-specific magnetization[6].In order to investigate the thickness dependence of the dichroism we did grow wedged shaped films with a typical slope of 2 ML/mm.We took then advantage of the small spot size (~50 mm) of the light. Growth, structural studies and SMOKE experiments have been performed with a different apparatus[7, 8].

In fig.1 we show the photoemission and difference spectra of the Fe 3p level for a 2 ML and 9 ML $Fe_{75}Ni_{25}$ alloy film.We can clearly see a different magnetic contrast between the 9 ML and 2 ML thick film. The thicker film has a dichroism of about 2% whereas for the thinner film one obtains a value of 10%. As discussed previously we can rule out changes in the growth mode or a reduction of T_c as the cause[3, 6].In fact RHEED studies have shown that the in-plane lattice constant contracts between 2 and 4 ML for $Fe_{80}Ni_{20}$ alloy films[3]. This suggests a structural driven effect.We therefore

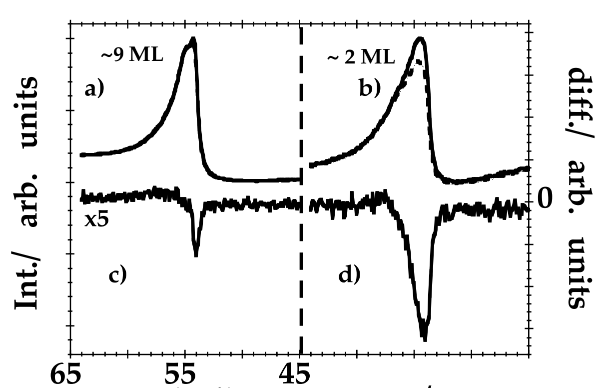

Fig.1: Photoemission (a)+b)) and difference spectra (c)+d)) for $Fe_{75}Ni_{25}$ films. Spectra a) and b) have been scaled to give the same peak height.The difference spectrum c) has been multiplied by a factor of 5.

performed LEED I-V studies to determine the perpendicular lattice constant. In fig.2 we compare the LEED I-V curves of the beam averaged (1,1) spot for 2 ML and 6 ML thick $Fe_{87}Ni_{13}$ alloy films.We can clearly observe that the peaks for the 6 ML film are shifted towards higher energies. This means that the perpendicular lattice constant is reduced. Together with the results of the RHEED experiments we conclude that the atomic volume is reduced by ~3%.

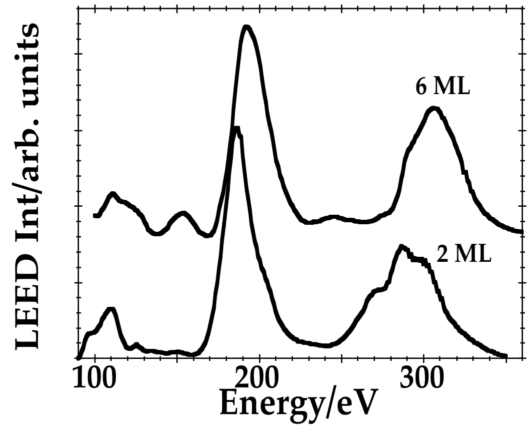

Fig.2: Comparison between LEED I-V curves of the (1,1) spot for a 2 ML and 6 ML thick $Fe_{87}Ni_{13}$ alloy film.

At this point it is important to stress that our films are not truly fcc, but tetragonal distorted. However up to this point we do not know how important the broken symmetry is. For example it is known for Fe/Cu(100) that the ferromagnetic phase is tetragonal distorted [9], which is different from the Fe/Cu(111) case in which ferromagnetism occurs in the isotropic fcc lattice [10].

In order to demonstrate that this is not a surface effect similar to 'magnetic live surface layers'[11] we show the thickness dependence of the SMOKE intensity for a $Fe_{87}Ni_{13}$ in fig.3. We can see that for 4-10 ML a linear increase of the SMOKE intensity is observed. Furthermore a linear curve through the data intercepts close to the origin. This means that the whole of the film is magnetic.

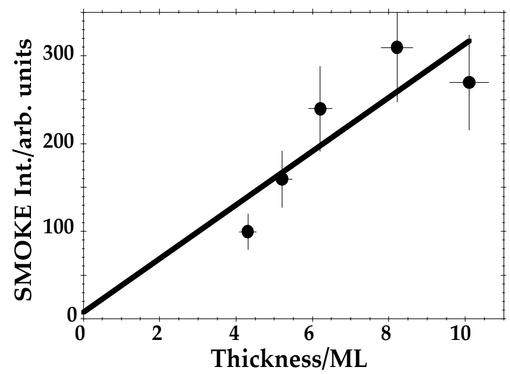

Fig.3: Thickness dependence of the SMOKE intensity for a $Fe_{87}Ni_{1}3$ alloy film.

Abrikosov et al. have calculated that in bulk fcc Fe_xNi_{1-x} alloys a moment collapse still occurs accompanied by a reduction of the atomic volume[2]. However they predict variations of the order ~9%, which is significantly smaller than our value of ~3%. Our value is also smaller than the change observed for Fe/Cu(100)[9]. Also in contrast with their calculation we do observe a ferromagnetic response in the form of hysteresis loops and magnetic dichroism.

It is now established that fcc Fe/Cu(100) exists in two phases with different atomic volumes ; the larger volume refers to a ferromagnetic high spin (HS) state and the smaller to an antiferromagnetic low spin (LS) state[12, 13] in agreement with theory [14].

The results of Keavney et al. suggest that a coexistence of phases is a possibility. They observed that by increasing the lattice constant of the substrate the average moment increases from 0.3-2.0 m_B[13]. However they found that the hyperfine field is not systematically dependent on the lattice constant. They described this as a change in the population of two coexisting phases. We would like to point out that we are not able to detect any antiferromagnetic order.Therefore we can not rule out the coexistence of antiferromagnetism and ferromagentism. Our results clearly show that additional magnetic phases other than a non-magnetic (NM) and high-spin ferromagnetic HS phases must exist for fcc Fe_xNi_{1-x} alloys.This follows from the observation of smaller changes in the atomic volume as predicted by Abrikosov et al. In general the formation of a moment requires an increased atomic volume.

We have observed a close connection between the Fe 3p asymmetry and the atomic volume. The observed reduction of the atomic volume at ~60-80% Fe content is significantly smaller than predicted by Abrikosov et al[2].

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